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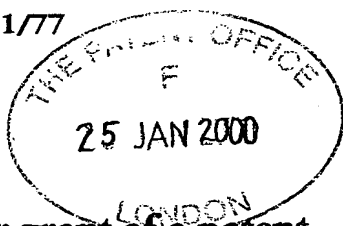
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4. Title of the invention

CATALYTIC BED REACTOR

5. Name of your agent (if you have one)

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Number of earlier application

Date of filing
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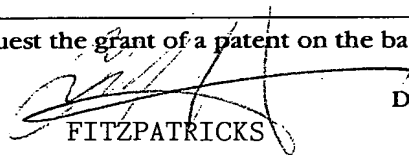
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CATALYTIC BED REACTOR

Field of the Invention

The present invention is applicable in the field of industrial chemistry and especially relates to improvements in chemical reactors. In general the invention relates to process control of temperature in a chemical reaction system and processing plant. Particularly the invention provides a reactor suitable for rapid conversion of a fluid reactant within the reactor in which the reactant temperature is maintained at a desired profile indirectly by means of a heat exchanging fluid.

Background of the Invention

In the majority of chemical processes there is a heat demand or a need to dissipate heat. Therefore, a wide range of chemical plant is involved in containing or conveying fluids which must at some stage of the process be either heated or cooled. One might consider furnaces, evaporators, distillation units, dryers and reaction vessels as plant where heat transfer manifests itself as a design and operational problem. In particular many industrial chemical processes employ reactors in which reactions are effected under given temperature and pressure conditions in the presence of a catalyst. Almost all these reactions generate or absorb heat i.e. they are exothermic or endothermic. The cooling effects for endothermic reactions generally adversely affect the rate of reaction and the corresponding parameters such as conversion and selectivity of the products from the reaction. The uncontrolled heating of exothermic

reactions generally leads to damage to the associated apparatus as the temperature can rise to a very high level. The reaction in such a case may become uncontrolled (so-called "run away reaction") and lead to
5 unwanted by-products and undesired effects, such as deactivation of a process catalyst. Furthermore, whilst an ideal catalyst does not theoretically participate in a reaction in reality many catalysts become degraded or poisoned as the reaction progresses and on an industrial
10 scale the costs associated with catalyst re-generation or replacement represent a significant burden. It will be understood that such costs also must include the down time for the plant or restrictions on capacity if a particular reactor has to be off-line for catalyst re-
15 generation purposes. Therefore, it is desirable to prolong the life of a catalyst bed in view of the significant cost benefits that may be obtained overall. The invention to be described hereinafter is ideally suited to use in catalytic reactor design but can be adapted for other purposes. No distinction is made as to
20 the application thereof to batch or continuous reaction systems.

Those skilled in the art recognise that it is beneficial for the changes in temperature resulting from
25 the heating or cooling effects of the reactions to be controlled. It is well known that maintaining the temperature of the reaction at a given constant level may result in significant advantages to the reaction, such as improved conversion and selectivity, prolonged life of
30 the catalyst and associated apparatus, reduced levels of unwanted by-products etc. In some cases, a slight

varying of the theoretical temperature profile may be more beneficial.

In order to effectively control the temperature of reactions within an acceptable range, the chemical industry has devised several arrangements, those commonly used being discussed in standard references and texts e.g. one might consider the general teachings by Octave LEVENSPIEL in Chapter 19 of Chemical Reaction Engineering. The relative merits of each approach is also discussed therein.

Conventionally, the temperature inside reactors has been controlled by passing an auxiliary heat exchanging fluid through tubes or between plates, same forming a heat transfer conductive medium or thermal bridge whilst separating the reaction species from the auxiliary heat exchange fluid. Thus it will be understood that in such an indirect heat transfer system there is on the one hand a process path or zone and on the other an auxiliary fluid path or zone separated by the tube wall(s) or plate surface(s).

Considering this well known concept in relation to packed catalytic bed reactors, reactant fluid is passed through the catalyst bed and heat of reaction therein is controlled by contacting the catalytic bed reaction zone with such auxiliary fluid containing tubes or plates. However, particularly for highly exothermic reactions, such an approach has not been found to be ideal since the packed bed often develops heat gradients, e.g. the catalyst bed will be cooler at its areas of contact with the said tubes or plates and hotter within its depth remote from said tubes or plates, permitting formation of

hot spots or moving hot fronts leading to variations in reaction progress within the bed as a whole. Thus at such a hot spot the reaction may proceed faster and hence catalyst therein will be more rapidly degraded. This
5 will be particularly significant in the case of large plants.

An example of an arrangement of a reactor designed to offer more control over the reactant temperature is the staged adiabatic packed bed reactor. This system
10 uses an arrangement wherein a number of discrete, spaced apart zones of reaction are provided with means therebetween to control the temperature of the products leaving a first zone of reaction prior to entering the next reaction zone. No heat exchanging means is provided
15 to control the temperature of the reaction in the zones of the reaction. Thus the reactant fluid entering the reactor at a desired temperature passes through a packed bed containing catalyst. Upon exiting this first stage, the reactant gas and any products will have a temperature
20 higher or lower than that of the initial temperature depending upon the reaction thermal characteristics. A heat exchanger then heats or cools the reactant gas to a second desired temperature, which may or may not be equivalent to the temperature of the first, before
25 passing to the next packed bed i.e. the second stage. This sequence is repeated until the desired conversion is obtained. Thus the temperature profile of the reaction will be stepped within an acceptable range of temperature, and will therefore not be truly isothermal
30 which is theoretically preferred.

An alternative proposal for a process and apparatus for controlling reaction temperatures is disclosed in US patent No. 5,600,053. This arrangement uses corrugated heat exchange plates spaced apart with each plate
5 defining a boundary of a heat exchange flow channel on one side of the plate and a boundary of a reaction flow channel on the other. In the arrangement, a heat exchange fluid passes in the first of the aforementioned channels and a reactant stream passes through the second,
10 preferably with a catalyst being present. This arrangement is intended to eliminate or minimise the typical step-wise approach to the so-called isothermal condition objective.

However, the arrangement proposed in US-A-5,600,053
15 requires adjacent corrugated plates to be joined together. For this purpose, smooth edges are provided to facilitate the assembly of superposed multiple plates to form channels. The plates are joined, such as by welding, along these smooth edges and hence the integrity
20 of the seal of the channels formed by the corrugations in adjacent plates is not ideal, particularly where a large pressure differential exists between the heat exchange flow channels and the reaction flow channels since this will tend to urge the adjacent plates apart. This
25 arrangement will thus place unnecessary constraints on parameters of the reaction, namely the relationship between the pressure of the heat exchanging fluid and that of the reactant gas.

An earlier system is described in US-A-5,073,352
30 which proposes an apparatus for conducting a process of reforming gasolines, under low pressure and in the

presence of at least one catalyst, in which heat required for the reaction is provided by a heat carrying fluid such as natural gas.

The apparatus described therein comprises a number of discrete reaction cells being arranged vertically and being of substantially parallelepipedic configuration. The cells are laterally spaced apart, thus forming channels therebetween for flow of the heat carrying fluid. The reforming catalyst-containing chambers are respectively either isothermal or adiabatic and dimensioned such that height (H), width (W) and thickness (T) satisfy the conditions $H > W > T$, and H is at least twice the value of W, W lying in the range of 50 mm to 10,000 mm (0.05 - 10 metres) and T lies in the range of 2 mm to 2,000 mm (0.002 - 2 metres). Thus there remains the possibility of hot spots and less than satisfactory thermal control in such large catalytic reactor volumes.

It is known to the man skilled in the art that the heat transfer coefficient in a packed bed is mainly dependent upon the catalyst particle size and the reactant fluid velocity through the catalytic bed. Unfortunately, both these parameters are process requirements and hence cannot be changed in order to improve the heat transfer coefficient in the packed bed, and hence in the reaction cells described in US 5,073,352. Additionally, it is difficult to move catalyst between narrow gaps or tubes, imposing limits on the dimensions of gaps or tubes through which catalyst is designed to flow.

Thus reactors of the known types according to the existing art have many significant limitations imposed on

the heat transfer capability. Plate reactors offer some advantages over tubular reactors on the auxiliary medium side, but the end result is not significant since the overall heat transfer coefficient is generally governed
5 by the process side as discussed above. Tubular reactors on the other hand offer advantages over plate reactors with regard to mechanical capability, due to increased resistance to differential pressure between the reactant fluid and the heat exchanging fluid.

10 Considering the foregoing matters, it is an object of this invention to provide improvements in chemical plant design and methods of operation thereof with a view to obviating or mitigating the drawbacks of the existing or previously proposed designs and methods.

15 Particularly, it is an aim of the present invention to provide an apparatus and a process for the control of reaction temperature within an acceptable range during operation of the chemical process by an indirect heat transfer method using a heat exchanging fluid.

20 Another object of the invention to be described more particularly hereinbelow is to provide an apparatus permitting control of the reactant temperature closely within a desired profile, more specifically, aiming to maintain the temperature at a substantially constant
25 level i.e. to offer attainment of an isothermal reaction zone in so far as is practical on an industrial scale.

It is a further object of the invention to provide chemical plant which is improved over known plant equipment in terms of both cost and space efficiency
30 considerations.

Summary of the Invention

The invention addresses the problems observed in the prior art by adopting the approach of staged adiabatic reactors and improving the performance thereof by design of a catalytic reactor comprising a catalyst bed and heat
5 exchange means of the plate type in operative contact with the catalyst bed, wherein the heat exchange means is formed from a plurality of superposed metal plates wherein fluid flow channels have been chemically milled
10 e.g. by etching, according to a pre-determined pattern, said channel-bearing plates being aligned during superposition to define discrete heat exchange pathways for fluids and diffusion bonded together.

The invention further addresses the drawbacks of the
15 known art by providing a process for conversion of a fluid reactant which process according to the invention uses a catalytic reactor comprising catalyst bed and heat exchange means of the plate type in operative contact with said bed and having discrete fluid pathways for heat
20 exchange between fluids at differing temperatures whilst avoiding mixing of the fluids, the said process providing the appropriate fluid reactant species to be converted in a catalytic reaction zone in the catalyst bed within the reactor and at a predetermined stage of reaction
25 introducing at least a portion of the fluid reactant species into a reactant fluid pathway within said heat exchange means, and also introducing an auxiliary fluid at a temperature differing from that of the fluid reactant species into another fluid pathway within said
30 heat exchange means and juxtaposed to the first whereby the discrete nature of the respective pathways permits

indirect heat exchange between the fluid reactant species, said process being optionally repeated in successive stages.

5 The residence time of the fluid reactant species within the catalytic reaction zone before introduction of at least a portion of same into the heat exchange means will be predetermined according to predicted reaction conditions and will be monitored in the usual way by observations made by a process operative or automatic
10 device sensing of reaction conditions including pressure and particularly temperature, such observations or sensing permitting adjustment of the process to advance or retard the progress of the reactant fluid as is considered necessary.

15 According to a modification of the process additional fluid reactant species may be introduced at subsequent catalyst bed stages. Thus although it is envisaged that the process can be operated in a series of stages in the manner of the known staged adiabatic
20 reactor systems the proposed reactor design permits greater control over the process not only in terms of heat management but also in terms of chemical reaction control.

25 Thus according to one aspect of the present invention, there is provided an apparatus for controlling the temperature profile of a reactant fluid in the presence of a catalyst during an endothermic or exothermic chemical reaction, comprising a reactor having reactant fluid inlet means and reactant fluid outlet
30 means; catalytic beds being provided therebetween, spaced apart by a heat exchanger; said heat exchanger comprising

heat exchanging fluid inlet means, heat exchanging fluid outlet means, a first channel or set of channels for passage of the heat exchanging fluid, and a second channel or set of channels in communication with the adjacent catalytic beds to allow passage of the reactant fluid from one catalytic bed to the next, said second channel or set of channels not being in communication with the reactant fluid.

Preferably a screen made of a fine mesh lines the walls of the catalytic bed, and said walls are ideally formed at least in part by plates of the said heat exchangers. The mesh acts to resist migration of catalyst into the reactant fluid-receiving channels of the heat exchanger down stream of the catalytic reaction zone.

The heat exchanger of choice is one formed from a plurality of plates superposed and diffusion bonded to form a stack of plates, wherein fluid channels are defined in said stack by virtue of a pre-treatment of said plates wherein each plate is selectively configured according to the desired pattern of channels by a chemical treatment to remove surface material e.g. by chemical etching, to a desired depth. Such a pre-treatment of the plates is conducted in a manner analogous to manufacture of printed circuit boards (PCBs) and for this reason the reactor design described herein can be described as a printed circuit reactor (PCR). The proposed reactor design offers an infinite variety of auxiliary heat exchanging fluid and reactant fluid pathways of very small dimensions which enables significantly enhanced process control.

The or each heat exchanger stack may be formed from a length or block of superposed plates by division e.g. by cutting into individual slices of a desired dimension which enables very slim designs of great strength.

5 Thus in one construction, a first such channel or set of channels is perpendicular to a second such channel or set of channels. In an alternative construction, the respective channels are parallel. Naturally one would generally arrange the construction such that juxtaposed
10 channels contain respectively reactant fluid species on the one hand and auxiliary fluid on the other to achieve the desired heat transfer. In this way temperature control is achieved indirectly without mixing of the reactant fluids with the auxiliary fluid media.

15 The profile of the channels, i.e. cross-sectional profile perpendicular to the flow path, is generally not critical but curved shapes are conventionally used and are relatively easy to form by chemical milling but other profiles could be adopted if desired by use of an
20 appropriate tool in combination with the chemical process.

In use of such a PCR reactor, the heat exchanging fluid may be caused to flow in a direction substantially perpendicular to the flow of the reactant stream.
25 Alternatively, the directions of flow may be substantially parallel and either co or counter current according to the operators choice taking account of the reaction process to be controlled.

The invention is primarily intended for use with
30 reactions using heterogeneous catalyst systems.

Depending on the particular reaction, optimisation thereof may be obtained by providing more than one catalyst, and in particular by providing different catalysts in separate catalytic beds.

5 The heat exchanging fluid may be a liquid or gas, as is understood by those in this art. Such fluids typically include molten salts, molten metals or hot water to provide liquid auxiliary media or may be hot gases, steam or superheated steam whereby heat may be
10 indirectly added to a reaction system. In the converse case chilled liquids or gases may be used. Chemical engineers will readily consider the wide range of auxiliary working fluids available and be aware of both sensible heat and latent heat considerations in matching
15 the auxiliary fluid needed to the process demands of the reaction for conversion of the fluid reactant species.

Each heat exchanger stack or each auxiliary fluid media channel, or set of channels, may contain a different auxiliary fluid to optimise the temperature
20 profile of the reactant fluid within the reactor.

Operation of the PCR is preferably such that the temperature of the fluid is below 800°C and the pressure below 500 bars.

According to another aspect of the invention, there
25 is provided additional means to make the aforementioned reactor specifically suitable for use as a moving bed reactor, namely catalyst inlet means, catalyst outlet means and means for feeding new or regenerated catalyst into the catalyst inlet means, and further means to
30 remove catalyst from the catalyst outlet means.

Preferably catalyst is allowed to progress towards the catalyst outlet under the influence of gravity. The man skilled in the art will be aware of alternative manners in which moving bed reactors may operate and the scope of the invention should not be restricted to the particular method described herein.

In the case of such a moving bed reactor, the bed width is a multiple of the catalyst diameter preferably at least 3 times that diameter.

According to a still further aspect of the invention, there is provided a process for indirectly controlling the temperature profile of a reaction fluid in the presence of a catalyst during an endothermic or exothermic chemical reaction, comprising passing a reactant fluid from a reactant fluid inlet means in a reactor to a first catalytic bed before passing through a first channel or set of channels in a heat exchanger and subsequently passing to a second catalytic bed; passing a heat exchanging fluid from a heat exchanging inlet means to a heat exchanging outlet means through a second channel or set of channels in the heat exchanger; and exchanging heat between the heat exchanging fluid and the reactant fluid whilst passing through the heat exchanger; the products of the reaction leaving the last catalytic bed being passed to a reaction fluid outlet means.

In an alternative embodiment, the process for controlling the temperature profile of a reaction fluid in the presence of a catalyst during an endothermic or exothermic reaction further comprises passing catalyst through the catalytic bed, catalyst leaving the bed being replaced by new or regenerated catalyst.

Préferably the catalyst is a heterogeneous catalyst.

In a preferred embodiment, the heat exchanging fluid flows in a direction substantially perpendicular to the flow of the reactant stream. Alternatively, the
5 directions of flow may be substantially parallel and either co or counter current.

The heat exchanging fluid may be liquid or gas, in accordance with the prior art. Such fluids include molten salts and boiling water. Preferably the
10 temperature of the fluid is below 800°C and the pressure below 500 bars.

It will be recognised that the invention in its broadest aspect provides a staged reaction system containing successive chemical reaction zones and heat
15 transfer zones, the latter containing plate heat transfer surfaces bearing micro-channels etched therein according to a pre-determined pattern, and offering not only the optimum indirect heat transfer strategy but also the ability to individually prepare fluid reactants thermally
20 for the next reaction zone by selection of the inlet channel and its relationship to adjacent auxiliary fluid channels. The system can be specifically designed to handle substances of different volatilities.

Description of the Drawings

25 By way of example, the embodiments of the invention will be described with reference to the accompanying drawings:

Figure 1 is a sectional side view of a reactor in accordance with the present invention;

Figures 2A and 2B show the temperature profile for an endothermic reaction in an adiabatic reactor and in a reactor in accordance with the present invention, respectively; and

5 Figures 3A and 3B show the temperature profile for an exothermic reaction obtained through a tubular reactor cooled with an auxiliary medium and a reactor in accordance with the present invention, respectively.

10 Mode for Carrying out the Invention

Referring to the drawings, Figure 1 shows a reactor 1 provided with a reactant fluid inlet 2 and a reactant fluid outlet 3 through which a reactant fluid to be processed is passed.

15 The reactor 1 comprises at least one catalytic bed 4 in communication with the reactant fluid inlet 2. The catalytic bed 4 is substantially vertical and the chamber walls defined therefor are preferably smooth. These smooth walls facilitate the movement of the catalyst
20 through the chamber where the apparatus is configured as a moving bed reactor.

Provided adjacent to the catalytic bed reactor 4 are heat exchangers 5. The heat exchangers 5 have at least two sets of channels formed therein, a first set of
25 channels provided for passage of the reactant fluid from a first catalytic bed 4 to a second, and a second set of channels through which the heat exchanging fluid flows.

An optional screen of a fine mesh suitably dimensioned according to the size of the catalyst

particles is provided in this embodiment to cover the ends of the reactant fluid channels adjacent to the catalytic beds to prevent migration of catalyst into said channels, especially the down stream channels with
5 respect to the catalyst to reduce the risk of blockages inhibiting flow.

Flow performance can be optimised and blockages decreased inside the channels, if the channel walls are smooth and free of crevices. The channels are formed by
10 joining individual plates together, said plates having channels formed in at least one surface, such as by a milling process. In order to ensure the crevice free design, it is thus important that any joining process used to join adjacent plates together does not interfere
15 with the machined channels. This therefore limits the acceptability of use of conventional welding processes and exposes the weakness of prior art proposals such as US-A-5,600,053. However, diffusion bonding processes, wherein the plates are placed under pressure and heated
20 close to the melting temperature of the plate metal thereby encouraging grain growth across the boundary, allows such a crevice-free design. This also enables the plates to be joined adjacent each of the channels, increasing the mechanical capabilities of the channels
25 and allowing greater pressure differentials between the pressure of the reactant fluid and the heat exchanging fluid. This design of heat exchanger has been proven by the designers of the proposed PCR since 1985 when Heatric first introduced its compact printed circuit heat
30 exchangers (PCHEs). The application of the diffusion bonding technique is now understood in the art.

The design of such heat exchangers also facilitates a greater rate of heat exchange in a given volume, reducing the space requirements for a given reaction. Thus incorporating them in a custom reactor design offers
5 hitherto unforeseen advantages.

Depending on the performance requirements of the reactor 1, additional catalytic beds 4 may be provided, spaced apart by additional heat exchangers 5. The final catalytic bed 4 in the series is in communication with
10 the reactant fluid outlet means 3 to enable the exit of the products of the reaction from the reactor 1.

In the alternative embodiment wherein the apparatus is adapted to be suitable for use as a moving bed reactor, catalyst outlet means (not shown) are provided
15 adjacent the lower end of each of the catalytic beds 4, through which the catalyst is urged by means of gravity. The catalyst leaving the catalytic beds 4 may be fed to a regenerator and then passed through catalyst inlet means (not shown) provided adjacent the top of each of the
20 catalytic beds 4. Alternatively, new catalyst material may be passed through the catalyst inlet means, consequent to exit of catalyst through the outlet means.

Thus the apparatus of the invention facilitates a series of adiabatic reactions, the temperature of the
25 reactant stream being altered between successive reactions to maintain the reaction temperature within an acceptable range and thus conform to the desired temperature profile.

Figures 2A and 2B relate to endothermic reactions.
30 Figure 2A shows the temperature profile for an adiabatic

reactor wherein the temperature decreases at a decreasing rate due to the cooling effects of the reaction.

Figure 2B shows a similar reaction performed in accordance with the present invention. An adiabatic
5 reaction takes place in the first catalytic bed, the size of said bed having been determined by the desired temperature of the reactant fluid leaving the first bed. The reactant fluid is then heated in the first exchanger
10 for the process up to a maximum value. The sequence is repeated with additional catalytic beds and heat exchangers until the desired conversion is obtained, the lower and upper values for the temperature range having been selected to optimise the process, said range being
15 as narrow as required.

Figures 3A and 3B relate to exothermic reactions wherein heat is generated by the reaction. Figure 3A shows the temperature profile of the reactant fluid obtained in a tubular reactor, cooled by an indirectly
20 heat exchanging fluid, such as molten salts. Similarly, Figure 3B shows the temperature profile of the reactant fluid for an exothermic reaction in a reactor in accordance with the present invention. Again, the reactant fluid enters a first catalytic bed wherein an
25 adiabatic reaction takes place, raising the temperature of the fluid to a maximum permitted value before cooling of the fluid in a first heat exchanger. The sequence is again repeated until the required conversion is obtained.

The number of heat exchangers is calculated in
30 accordance with methods known to the man skilled in the

art in order to avoid hotspots and to allow the process to run at a higher load of feed per unit of flow rate.

For example, phthalic-anhydride may be obtained by oxidation of ortho-xylene in the presence of air. For
5 such a reaction, conventional tubular reactors are hot spot limited, and as a consequence, the maximum amount of oxide in air is limited to about 80 g/Nm². Performing the same reaction in a reactor in accordance with the present invention allows much higher rates of reaction
10 and consequently an increased throughput.

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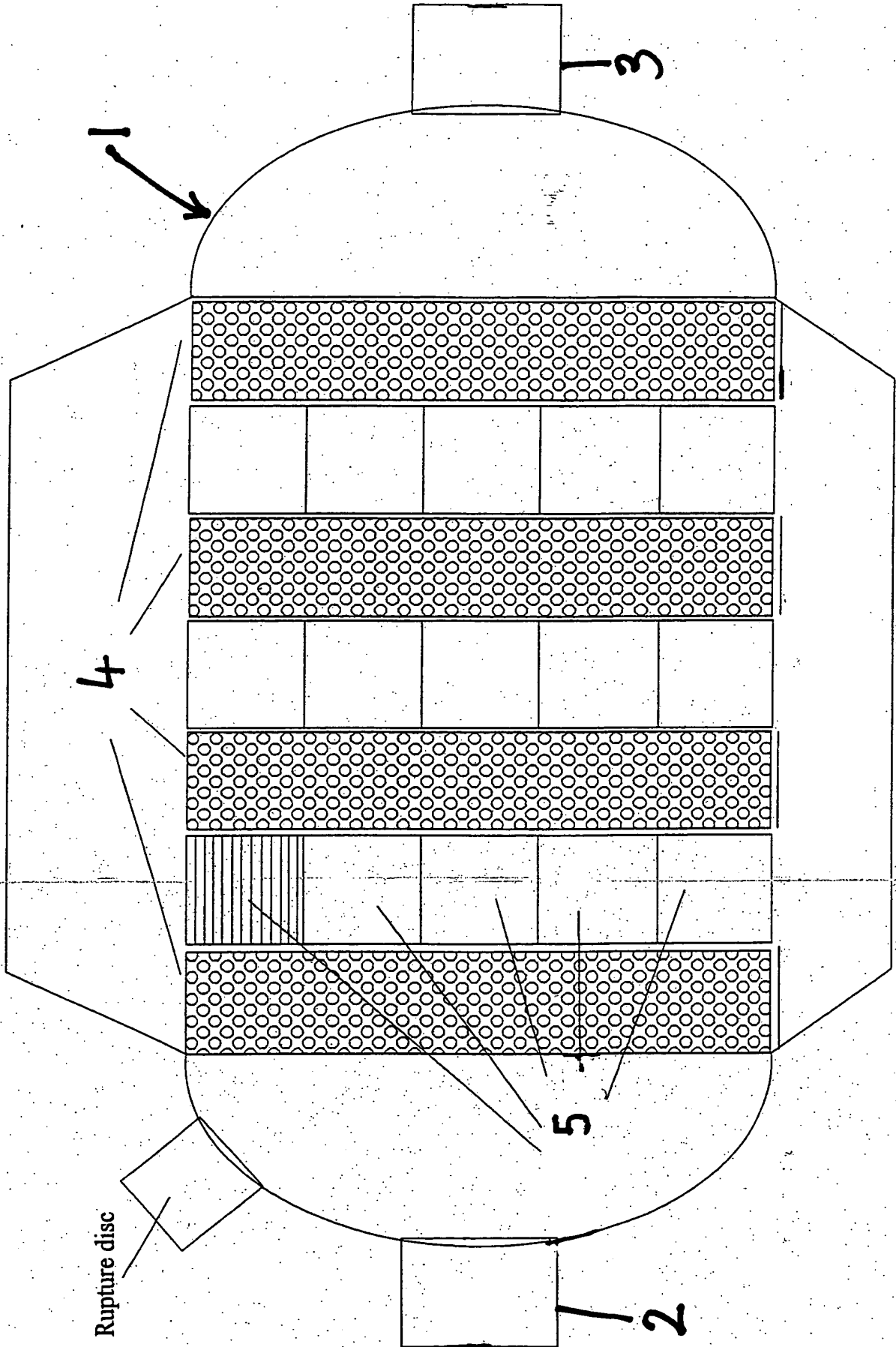


FIG 1

Temperature profile in an adiabatic reactor

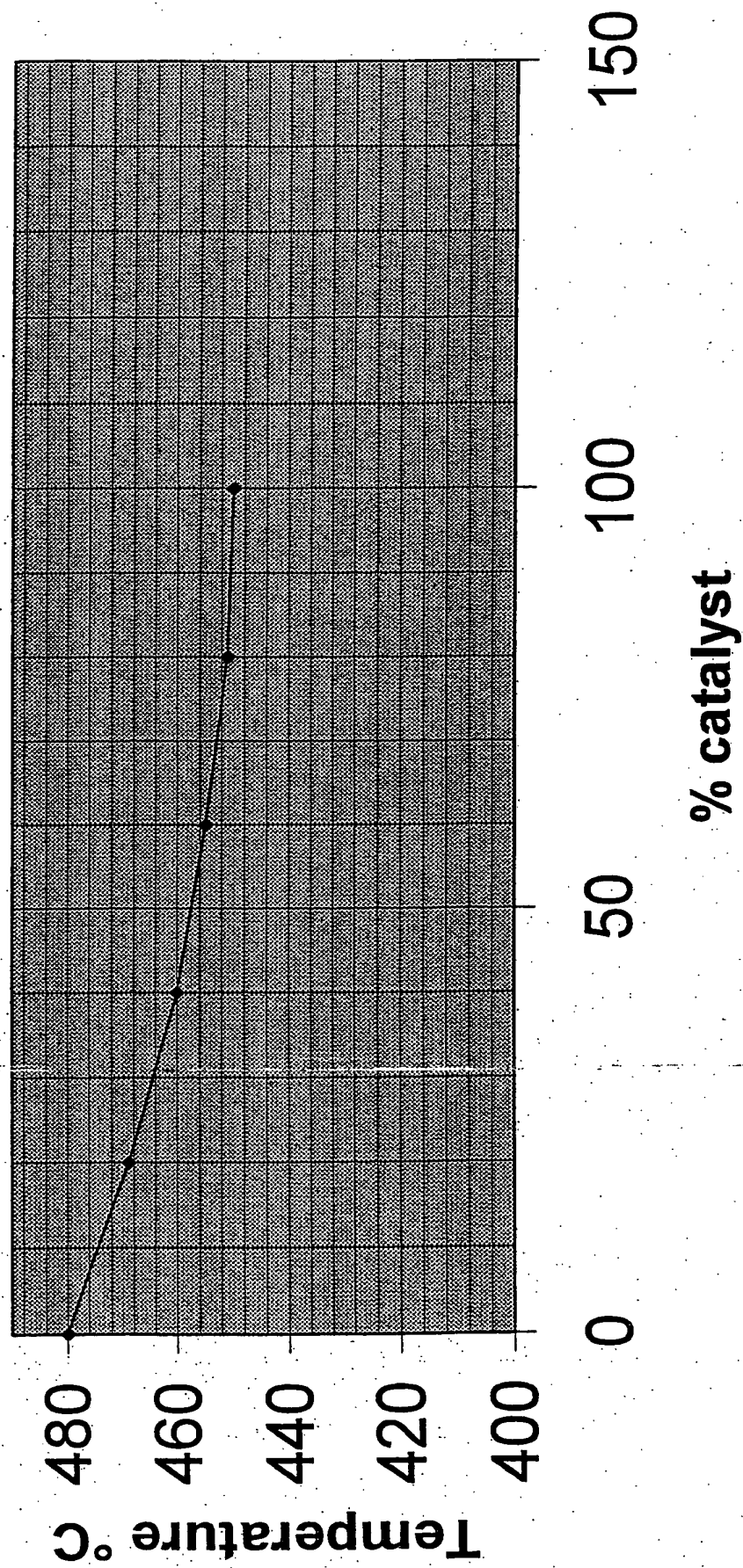


FIG 2.a

Temperature profile in a PCHE reactor using 3 exchangers

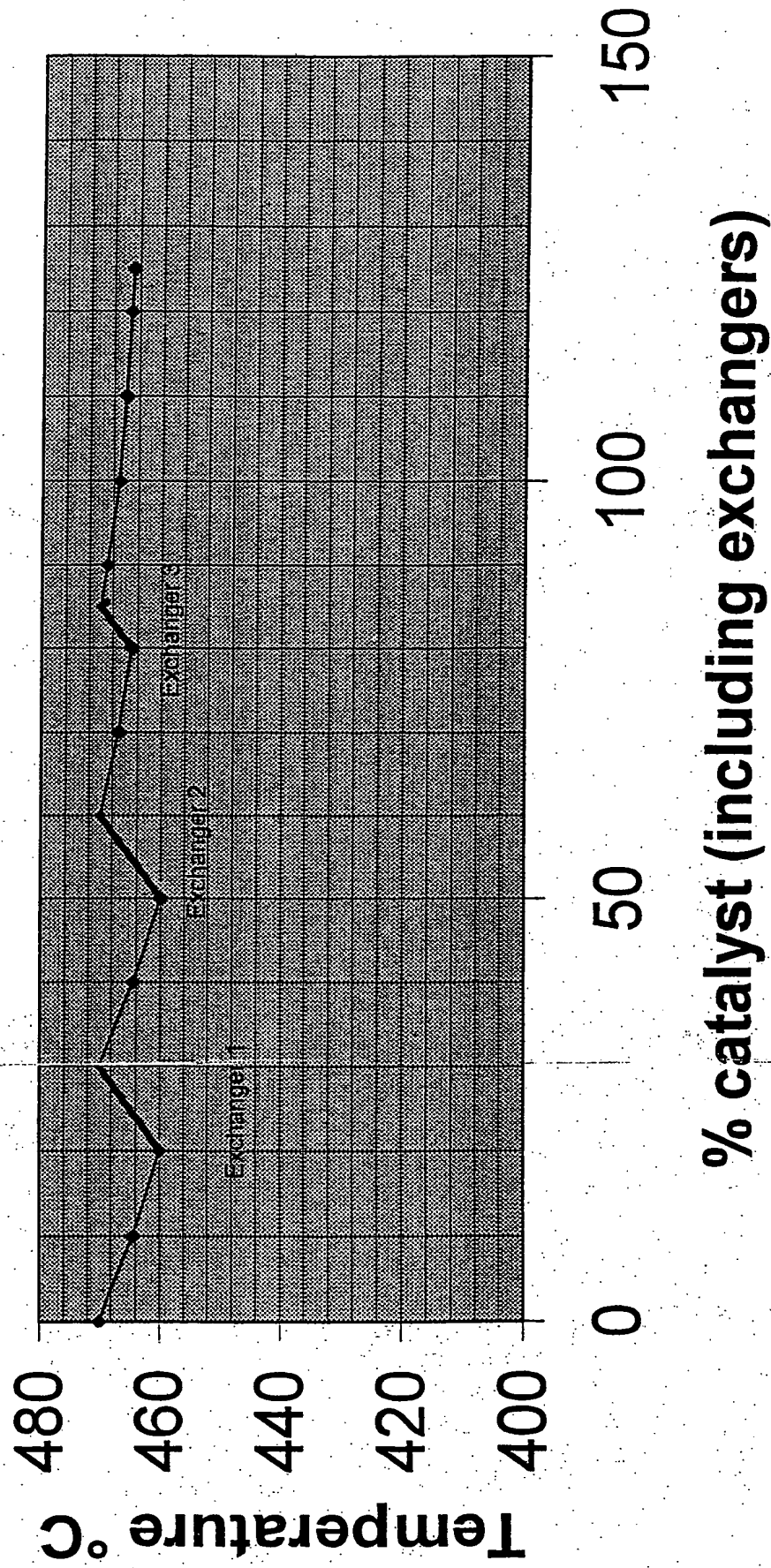


FIG 26.

Temperature profile in a tubular reactor for an exothermic reaction

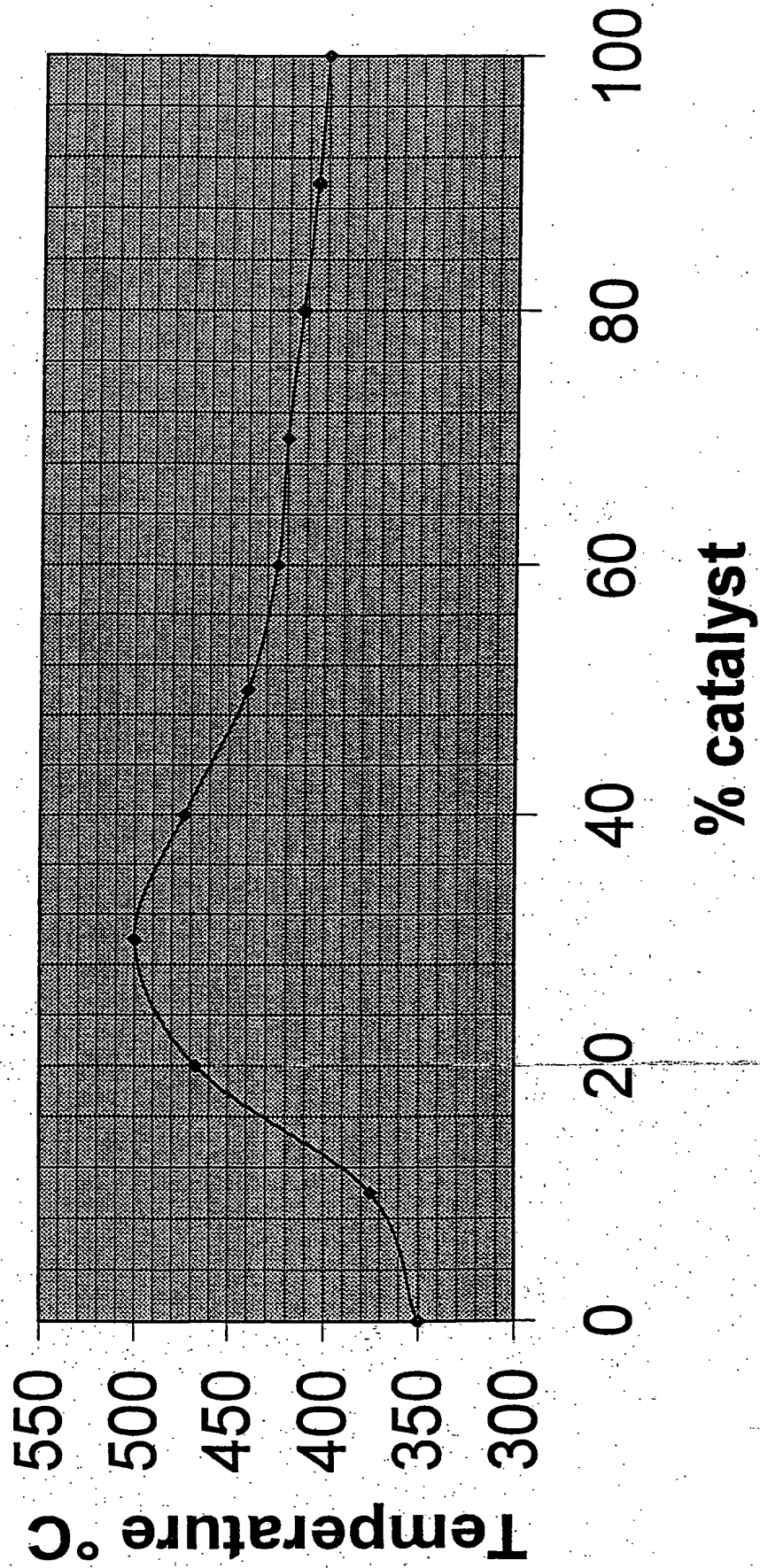


FIG 3a

Temperature profile in a PCHE reactor for an exothermic reaction using 5 exchangers

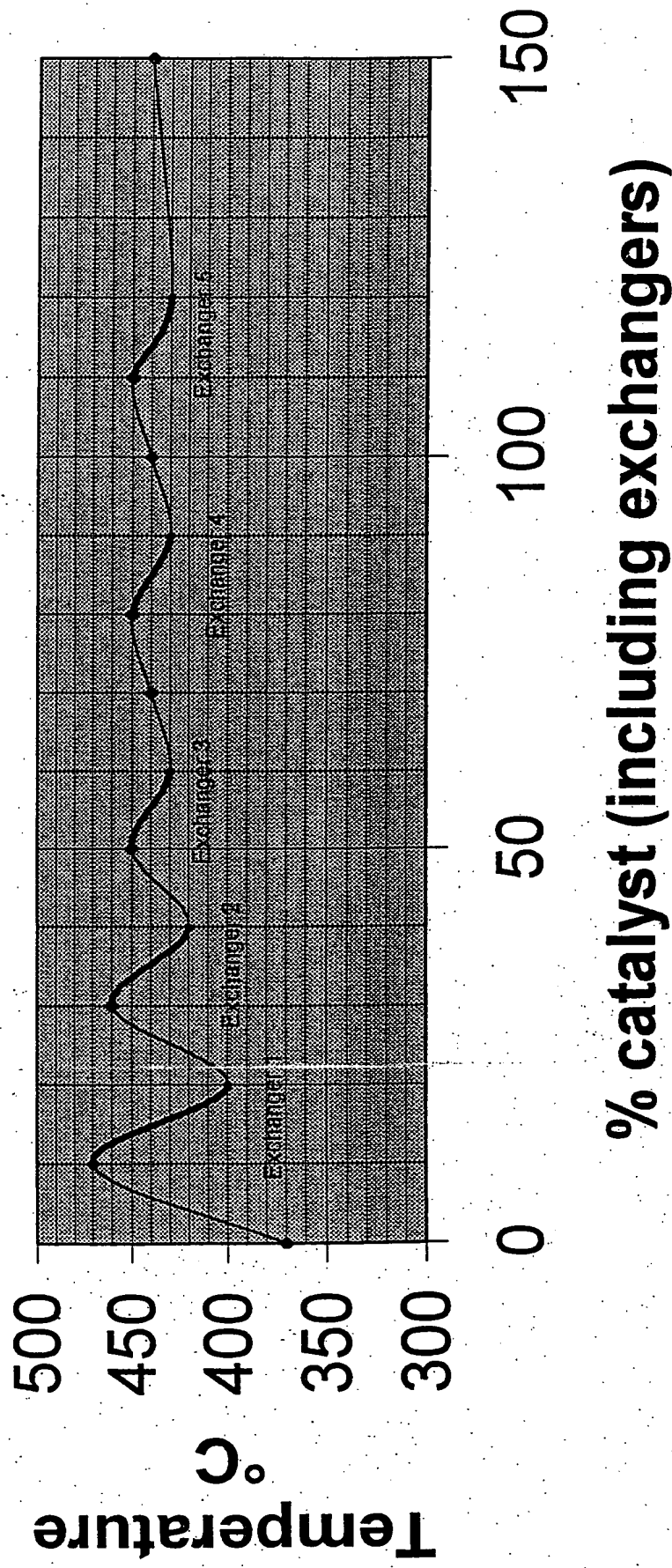


FIG 3.b

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